

***Independent Peer Review of  
Source Term Modeling for the  
INEEL Subsurface Disposal  
Area***

*Dr. Matt Kozak  
Dr. Man-Sung Yim  
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**Idaho  
Completion  
Project**

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# **Independent Peer Review of Source Term Modeling for the INEEL Subsurface Disposal Area**

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## **ABSTRACT**

On Friday, September 5, 2003, the Idaho National Engineering and Environmental Laboratory called a review meeting to examine source term modeling for the Subsurface Disposal Area. The review team, defined in the report, answered specific questions and provided general observations. The details of this review are what comprise this report.

# Independent Peer Review of Source Term Modeling for the INEEL Subsurface Disposal Area

## Background

On Friday September 5, 2003 a review meeting was called by INEEL to examine source term modeling for the Subsurface Disposal Area (SDA). The review team was comprised of Matt Kozak, Monitor Scientific, LLC, Terry Sullivan, Brookhaven National Laboratory, and Man-Sung Yim, North Carolina State University. INEEL staff present for the meeting included Steve Priebe, Jean Holdren, Bruce Becker, James McCarthy, and Roger Seitz.

The meeting began with a presentation by Jean Holdren describing the project background, current modeling approach, end use for the models, and objectives for the review team. She identified four major objectives:

- Review SDA source release modeling to assess appropriateness, strengths, and vulnerabilities for supporting remedial decisions.
- Recommend modifications for future analysis in the remedial investigation/baseline risk assessment and the feasibility study to evaluate long-term effectiveness of remedial alternatives.
- Recommend specific actions to assess sensitivity and uncertainty
- Recommend specific actions to reduce uncertainty.

Bruce Becker presented a more detailed description of the modeling approach. He covered the physical description of the 97-acre disposal area, and the modeling assumptions for inventory, infiltration, container performance, and waste form performance. During both presentations the review team asked a number of questions to help clarify the modeling approach and modeling needs for the SDA. Based on the information provided in the *Ancillary Basis for Risk Analysis of the Subsurface Disposal Area*, documents referenced therein, and these presentations the review team was asked to address seven questions geared towards achieving the four major objectives listed above.

## General Observations

The review team feels that in general the INEEL staff members are performing a credible and defensible analysis of the source term. The situation is complicated by the long-time history (disposal started in 1952) and lack of reliable information on inventory, physical and chemical form of the inventory, container performance, waste form release characteristics and transport parameters. The INEEL staff has worked hard to reduce the uncertainties associated with these areas. However, uncertainties still exist and one of the focuses of this review is on how to further reduce uncertainties.

For supporting feasibility studies where the objective is to evaluate effectiveness of remedial actions, it may be more desirable to use realistic values for parameters. Conservative modeling assumptions were applied in the absence of sufficient site-specific data, leading to the predicted release of a substantial fraction of the inventory prior to the start of source zone remediation. Thus, remediation would not be effective. Consequently, to properly assess the impacts of remedial alternatives more realistic modeling of release and transport is required.

## **Questions posed to the review team**

### **1. Is the DUST-MS model developed at Brookhaven National Laboratory (BNL) appropriate for application to the INEEL SDA?**

Yes. DUST-MS has the container performance and release models needed to perform the analysis. The current information in the source term models in DUST-MS is equal or better than other extant computer codes. A limitation in the DUST-MS model is treatment of solubility for multiple isotopes of the same radioelement (e.g., uranium). DUST-MS provides the capability for a user to specify solubility for each isotope, but does not check that the sum of the isotope concentrations exceeds the solubility limit for the isotope. This is potentially of concern for the next round of analyses, in which uranium solubility limitations will be applied to some waste forms.

### **2. Are there other readily adaptable models that would be superior for this particular application?**

No. There are other readily adaptable models but they are not necessarily superior to DUST-MS. Many of these models are proprietary (hence unavailable) or available only through licensing agreements. Some models have some advantages over DUST-MS (solubility of multiple isotopes) but will have limitations compared to DUST-MS (e.g., may not allow as many release mechanisms or a distribution in container failure time).

### **3. Do multiple applications of the one-dimensional DUST-MS model adequately represent the two- or three-dimensional source term release distribution in the SDA?**

Yes, provided an appropriate number of different source regions are used. Use of multiple one-dimensional source zones over the 97-acre disposal facility provides a quasi-three-dimensional representation of the source. Spreading of the plumes should be minimal in the few meters of the source zone depth, so mixing between disposal areas should not be extensive with the possible exception of the narrow trenches. However, these appear to be already lumped into a single source.

An important issue is how to define the source zones in an acceptable manner. Within disposal pits a number of high-activity sources (beryllium blocks, contact handled waste, etc.) can be found. A sensitivity analysis should be performed to determine the impact of these high-activity sources (hot spots). While it is clear that modeling only the high activity regions will lead to highly localized fluxes from the source zone, the scale of the hot spot may be small and mixing in the vadose zone may cause dilution of the hot spots. The objectives of the sensitivity analysis should be to define the number of sources to represent in the model.

### **4. Are assumptions and DUST-MS model parameters appropriate?**

Yes, in general. Specific issues and recommendations follow for model assumptions and parameters.

### ***Model Assumptions:***

One deficiency was noted in the DUST-MS code in response to Question 4 and it has to do with a unique solubility limit for each isotope. For species with multiple isotopes this is not correct as the solubility is a property of the element (i.e. fractional solubilities for each isotope sum to the total element solubility). Consequently, the impact on individual isotopes may change as a function of time, owing to differing decay rates of the isotopes. For elements such as uranium, careful attention should be placed on properly modeling solubility constraints. It may be possible to obtain a defensible estimate of release because large changes in the isotopic ratio during transport are not anticipated over the time scale of the analysis, but this should be carefully evaluated.

The release mechanisms applied by the INEEL team (dissolution for glass and metal, diffusion for concrete and for gases through sludge, and surface rinse for everything else) seem to be appropriate for the waste forms. The INEEL team is taking a reasonable amount of credit for container properties. Waste form properties for the most part have been chosen to over-predict actual release (e.g., diffusion coefficient of  $10^{-6}$  cm<sup>2</sup>/s for concrete wastes, instant release from fuel-type wastes). The analysis team does not use worst-case values for K<sub>d</sub>, but uses average values that are appropriate.

The corrosion rates for the container are based on site-specific data and conservatively estimate the time to failure.

### ***Parameters:***

#### **Inventory**

Time did not permit an in-depth review of the inventory estimates, but it appears that the INEEL staff has performed an extensive amount of work to reduce uncertainty and has done an excellent job compared to other programs involving historical waste. Most of the substantive results are based on best estimate of the inventory. This is consistent with attempting to provide reasonable estimates of potential impacts. Sensitivity analysis was performed to examine the impacts of uncertainties in inventory on predicted outcomes.

Tc-99 and I-129 are often problematic when performing assessments of radioactive waste. It is suggested that a review of the Tc-99 and I-129 scaling literature by Jene Vance (Vance 1992) should be performed to check the appropriateness of INEEL inventory estimates. Vance's work used commercial reactor data, and applied to inventory estimates for commercial waste from BWRs and PWRs. Special scaling factors may be needed for DOE and Naval Reactors. This may lead to reductions in Tc-99 and I-129 inventory by a few orders of magnitude. If the inventories were much lower, it could help explain the discrepancy between modeled and measured concentration, although other factors (waste form release or chemical interactions of Tc in the subsurface) could also be involved.

### **Infiltration**

The use of a spatially variable infiltration field leads to dispersion in the near field that will significantly lower the peak risks. This is demonstrated in Figure 6-91 of the Ancillary Basis for Risk Analysis. In addition, the approach to averaging infiltration for specific waste types (source zones) may damp out extremes in the infiltration rate. It was noted that in some source zones, the infiltration was bimodal: either 3.7 or 24 cm/year, yet a value of 11 cm/yr was used in the model for this zone.

Justification for the use of spatially variable infiltration should be improved through comparison to field data. In the absence of clear justification of the spatial distribution, a facility wide average should be used. Sensitivity analysis on the range of anticipated infiltration rates, as was done in the Ancillary Basis for Risk Analysis, should also be repeated for the baseline risk assessment.

Temporal variations in infiltration and variations in release will occur, but we do not believe that this is a significant issue provided a properly averaged yearly infiltration is used. Flooding has been observed in the past; however, berms have been installed and it is our understanding that this has alleviated the problem. If flooding is a credible occurrence in the future, sensitivity analysis of the impacts of flooding should be performed.

### **Waste form release characteristics**

The corrosion rates used for carbon steel waste forms appear to be too high. They are inconsistent with the container failure rates and should be checked. Romanoff studied corrosion of carbon steels and wrought iron for periods of up to 14 years and is a source of data for examining this issue (Romanoff 1957).

The use of site-specific data for drum failure rates is appropriate and commendable. The data support the values used in the assessment. Consideration should be given to comparing the site-specific data to literature data found in Romanoff (Romanoff, 1957).

Surface wash-off for fuel waste forms is conservative. Credit for dissolution can be claimed. Typically, a small fraction in the gap is released instantly and the remainder will be controlled by dissolution. Review of the Yucca Mountain literature on fuel performance is recommended.

Review of the Yucca Mountain cladding performance literature is recommended to determine release rates from cladding materials. This may have to be adjusted for the fuel-type wastes disposed of in the SDA.

### **Geochemistry: Sorption and solubility**

This area appears to be the weakest, but it is commensurate with the technical approach used in other source term modeling programs. The approach for sorption is based on site-specific data where available and on Sheppard and Thibault, a standard reference in the field, when data are unavailable. More recent literature on soil Kds should also be reviewed. See reference list below.

Use of a geometric mean value for Kd in the waste region based on soil Kd values may not be appropriate. The presence of organics, chelating agents, etc. may greatly reduce Kd. The

literature should be reviewed for the effects of organic materials that might lower Kds. See reference list below.

The use of a Kd of 22 for Pu was too low as judged by an independent review panel. (Berkey, Weirenga, and Roback, August 28, 2003). That review team felt a value of 5100 was appropriate based on the data.

As discussed previously, applying solubilities to uranium will lead to problems in the DUST-MS model.

A number of investigators have reviewed the literature on solubilities in cementitious wastewaters that will be useful for grouted wastes and for the presence of organics. See reference list below.

The possibility that there may be a small fraction (<1%) of Pu and other contaminants that are mobile should be investigated.

#### **Other**

Moisture content is an issue because it is not coupled to velocity in DUST-MS. Generally, calculated results are not very sensitive to moisture content and average values should be acceptable. This is particularly relevant for sorbing contaminants.

Bulk density of the wastes may not be the same as in the soils. The bulk density of the wastes is not expected to vary from the soils by more than a factor of two. A factor of two increase in bulk density would lead to approximately a factor of two increase in sorption and a reduction in solution concentration by a factor of 2. This is not viewed as a major difference in calculated outcomes.

The height of the facility (i.e., thickness of the waste zone) can impact the predicted concentrations for a fixed total inventory. Increasing the facility height by a factor of two would decrease the peak concentration by a factor of two for a fixed inventory. The release would also be spread over a longer period of time. Sensitivity analysis on the impacts of facility height may be valuable in assessing the impact, but this is likely to be less important than the geochemical effects.

#### **5. What assumptions, release mechanisms, and parameters should be modeled to evaluate sensitivity and uncertainty?**

See discussion following Question 4. Based on experience elsewhere, the parameters that are expected to have the highest impact on release are solubility, sorption, infiltration, and standard deviation on the container failure rate distribution. Changes in the release mechanism can also substantially impact release (e.g., changing from surface rinse to dissolution as suggested for fuel-type wastes). Changes in waste form release parameters for the dissolution or diffusion models can also impact release to some degree (e.g., reducing diffusion coefficient or dissolution coefficient by orders of magnitude, which only could be done with new data to support the changes).



For contaminants that can exist in the gas and liquid phase care must be taken in modeling. Conservative assumptions for one-phase may be non-conservative for the other phase. Exchange of inorganic C-14 between soil gas and groundwater can take place through a complex mass transfer process. Assuming a groundwater source, this transfer can release C-14 to the atmosphere, contributing to the depletion of C-14 in the groundwater. The mass transfer coefficient of CO<sub>2</sub> was measured between soil gas and a simulated groundwater in static soil columns (Caron et al. 1994), and in a pilot-scale experiment with a moving aquifer (Caron et al. 1996; 1998a). This work on C-14 partitioning should be reviewed.

**6. What assumptions, release mechanisms, and parameters should be modelled or measured to evaluate sensitivity and uncertainty?**

Environmental monitoring cannot be expected to provide adequate feedback to reduce uncertainty in the source term release, owing to the difference in time scales between the measurements possible in monitoring and the time scales of concern for performance assessment. Additionally, it may be difficult to separate uncertainties in transport to the monitoring location from the wastes to uncertainties in the source. To reduce source term uncertainty requires tests specifically designed to understand release characteristics. These could be laboratory –scale or field-scale tests. Field-scale tests would require measurements close to the source to eliminate other processes as being the cause for the observed behavior. Prior to conducting an experimental program, it is recommended that detailed sensitivity analysis be performed to identify parameters that lead to the biggest change in predicted outcomes.

Specific areas that may lead to reduced uncertainties are:

- Improving K<sub>d</sub> estimates for the wastes would require measurements on real wastes. The expense and worker exposure associated with collecting the K<sub>d</sub> values may outweigh the potential benefits of the data. Even if data were available on wastes, questions pertaining to representativeness and other uncertainties would still exist.
- Examination of gas phase transport and partitioning between the aqueous and gas phase.
- Use of the geocentrifuge to examine issues pertaining to flow and transport processes in concrete or unsaturated soil should be explored.

**7. What modeling modifications should be adopted to address near-term (e.g., within three years), mid-term (e.g., during remedial design within five to seven years), and long-term (e.g., beyond seven years) evaluation and remediation.**

The review team feels that the existing models and approaches are adequate for addressing near-term future modeling issues. However, several relatively minor deficiencies (discussed above) in the DUST-MS code would be useful to address.

The review team did not address mid-term or long-term modeling issues for two reasons. First, the objectives for mid- and long-term modeling are not clear. The focus of this review was to examine source term modeling to support remedial design and decision making. Second, model and data needs for the mid- and long-term will depend on the selected remedy.

### **Recommendations**

The review team offers this list of recommendations to address potential areas for improvement. The improvements suggested are aimed at reducing uncertainties and increasing confidence in model results.

- Modifications of DUST-MS to deal with the solubility issue of multiple isotopes of the same radioelement.
- Modifications of DUST-MS to deal with time-dependent changes in material properties (moisture content or perhaps transport and release parameters if needed). For example, if grout is used there may be a long period of high pH, which returns to the natural state over several hundred years.
- To build confidence in the INEEL results, QA testing of the integrated code (DUST-MS, TETRAD, DOSTOMAN) against other available codes for simplified conditions. It is not possible to compare to the actual conditions based on the complexity of the situation. Such testing and benchmarking should be carried out against as complex a model as possible.
- To build confidence in DUST-MS, further QA testing may be needed. QA testing was performed for DUST-MS, however, it is not widely available. Improved documentation of existing testing of DUST-MS is appropriate.
- Improved abstraction of the unsaturated zone system of models to improve its computational efficiency would allow improved treatment of uncertainty/sensitivity of the total system model. The current model appears to be a very complex system of transient flow and transport in the unsaturated zone, which may be overly complex and detailed given the needs of the modeling.
- A clearly identified structure for balancing worker risks and other costs versus long-term risks is needed.
- If probabilistic analysis is performed, a post-processor to collect and manage the appropriate data from each run will be needed. Proper QA management will permit sensitivity analysis to be easily performed.

### **Other Suggestions**

As part of the review the following suggestions/observations are provided.

- For comparison of remedial alternatives (RI/FS studies), consider examining only the source zone. Once contaminants are released from the source zone the transport should remain unchanged for the various analyses unless major differences in transport parameters are envisioned (i.e., plume of high pH water from grout changing  $K_d$  and solubility limits). If it is acceptable to examine only the source zone, computational requirements decrease and sensitivity/uncertainty analysis or probabilistic analysis become much more tractable.
- The potential for analyzing the impacts to inadvertent intruders was mentioned during the meeting. There is no direct requirement in CERCLA for this type of analysis. However, there are requirements for institutional control and periodic re-evaluation of performance. Inadvertent intruder analysis is used after the institutional control period in performance assessments of radioactive waste disposal sites. For the Hanford Tanks and Oak Ridge Solid Waste Storage Area 6, inadvertent intruder analysis was performed. Inadvertent intruder analysis typically uses a higher dose limit than for the general public and is often used to set waste acceptance criteria. In fact, the waste classification system in NRC Part 61 is based on protection of inadvertent intruders. Since INEEL has no control over acceptance of past wastes, careful thought should be given to the impacts of this type of analysis on remedial goals.

#### **A List of References for the Project**

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## **Appendix A**



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October 7, 2003  
Dr. Steve Priebe  
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Idaho Falls, ID 83415-3875

Dear Steve:

Attached is the peer review report on "Source Term Modeling for the INEEL Subsurface Disposal Area," conducted by Drs. Kozak, Yim, and myself. If you have any questions please contact me.

For the committee, it has been our pleasure to perform this review and we appreciate the access provided to INEEL staff and information necessary for this review.

Best regards,

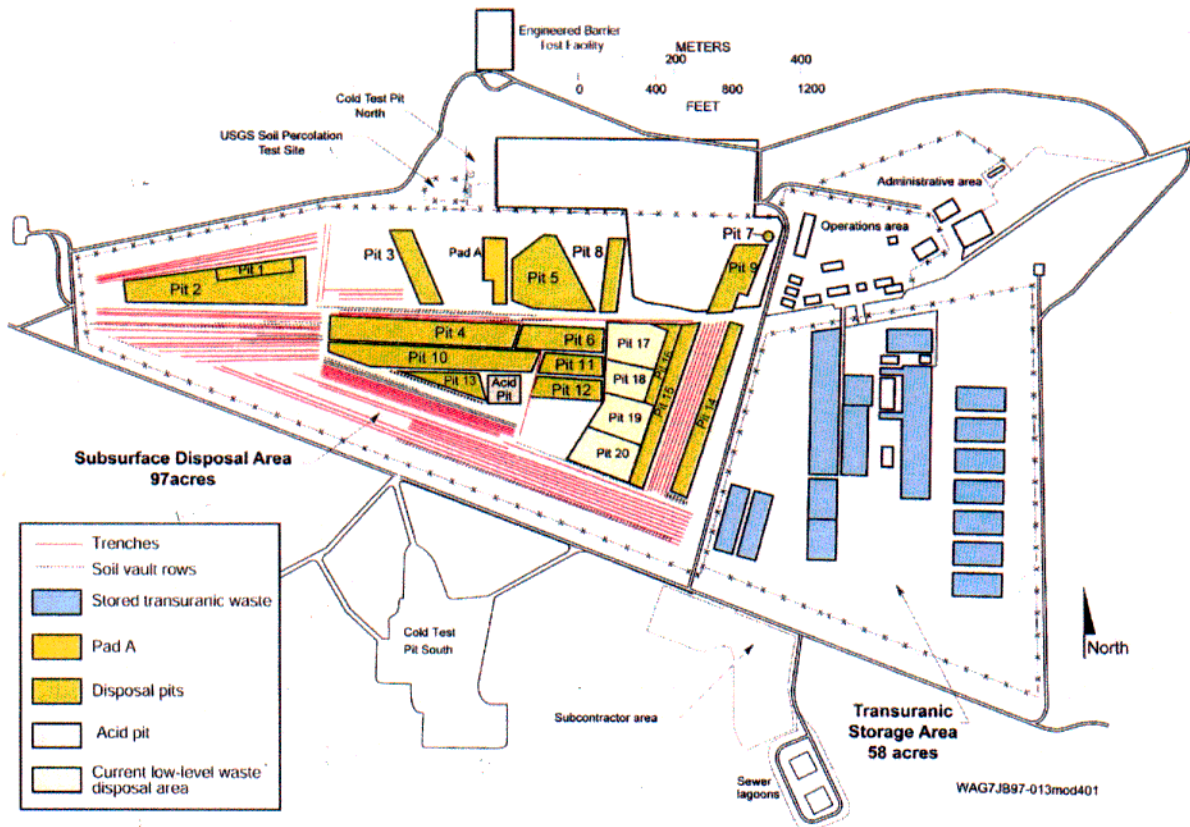
A handwritten signature in black ink that reads "Terry". The signature is written in a cursive, flowing style.

Terry Sullivan  
Environmental & Waste Technology Group

cc:



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Monitor Scientific, LLC



Terry Sullivan  
Brookhaven National Laboratory



Man-Sung Yim  
North Carolina State University

## **BIOGRAPHIES**

### **Matt Kozak**

Dr. Kozak is known both nationally and internationally for his work on safety assessments of waste disposal facilities and contaminated sites. Dr. Kozak is a frequent consultant to the International Atomic Energy Agency, and has supported the governments of Belarus, Bulgaria, Egypt, Estonia, Moldova, and Poland on missions to site, develop, construct, and analyze disposal facilities to provide national capacity to disposal of radioactive waste. He was the official U.S. delegate to the International Atomic Energy Agency's Coordinated Research Program on Improvement of Safety Assessment Methodologies (ISAM), and is currently participating in its successor program on Application of Safety Assessment Methodologies (ASAM). He has also conducted recent project work in Canada, the UK, Korea, Japan, and South Africa.

In the USA, Dr. Kozak has supported EPA, DOE, and NRC on a wide variety of radioactive waste disposal and radioactive contamination issues. This has included work on TSPA for Yucca Mountain, and a number of other sites for low-level waste. He has provided technical input to regulatory rulemakings for high-level and low-level wastes, and for residual contamination from decommissioning.

During 2000-2001, Dr. Kozak was a member of the National Research Council Committee on Cesium Processing Alternatives for High-Level Waste at the Savannah River Site. Dr. Kozak is member (and former chair) of Scientific Committee 87-3 for the National Council on Radiation Protection (NCRP) on safety assessment of radioactive waste disposal facilities. Dr. Kozak is also a member of NCRP Umbrella Scientific Committee 87 on Radioactive and Mixed Waste.

### **Terry Sullivan**

Dr. Sullivan is a member of the Environmental Research and Technology Division staff at Brookhaven National Laboratory. Dr. Sullivan joined BNL in 1983 and has gained national and international recognition for his work on source term assessment for near surface radioactive waste disposal. Dr. Sullivan's primary research interest is in the application and development of models for soil and groundwater contamination problems. For the Nuclear Regulatory Commission he has developed six different computer models to perform source term analysis and predict subsurface fate and transport from shallow land disposal facilities. These models have gained international acceptance and use. Dr. Sullivan has provided several International Atomic Energy Agency (IAEA) courses on low-level waste source term analysis and has been a technical expert for the IAEA on five missions. He is also an active member of the IAEA programs on Applied Safety Assessment Methodologies (ASAM) and Disposal of Decontamination Wastes. He is a member of the National Council on Radiation Protection's subcommittee on safety assessment of radioactive waste disposal facilities.

Dr. Sullivan's other research interests are in risk assessment and management and the use of decision support software to assist in defining clean-up goals in environmental remediation problems. He worked with the Department of Energy's Center for Risk Excellence on a wide range of risk related issues. He was the principal investigator, for the Environmental Protection Agency, in an Environmental Technology Verification study of decision support software. For DOE and EPA he has prepared 3 state of the art reviews for decision support software and has

organized and chaired a NATO session on this topic. Dr. Sullivan is currently the principal investigator for a program that examines human health risks associated with mercury deposition resulting from coal-fired power plants.

Dr. Sullivan has also been called upon to provide technical assistance for a wide range of environmental contamination problems. For the DOE he has led technical review teams for assessing remediation of contamination associated with the separations processing research unit at Knolls Atomic Power Laboratory, radionuclide and heavy metal groundwater contamination at the F and H area seepage basins at the Savannah River Site, and water treatment options to remove uranium from process waters at the Fernald site. He has also been a member of review teams that examined organic and radionuclide groundwater contamination issues at the Lawrence Berkeley National Laboratory and research and development needs at Savannah River Site and Idaho National Engineering Laboratory. At Brookhaven, he has provided technical support on groundwater and river sediment contamination issues.

#### **Man-Sung Yim**

Dr. Yim is Associate Professor and Director of Graduate Programs of the Department of Nuclear Engineering at North Carolina State University and is currently a Sam Nunn Security Fellow at Georgia Tech. He is also Technical Coordinator of Department of Energy Office of Civilian Radioactive Waste Management Fellowship at North Carolina State University. He performs research in nuclear waste management, risk analysis, and advanced nuclear fuel cycle analysis and teaches graduate level courses on nuclear waste management, radiation safety and shielding, radiological assessment, and environmental exposure and risk analysis. Prior to joining NCSU Nuclear Engineering Department in 1995, he was a lecturer at MIT (Department of Nuclear Engineering), senior researcher/researcher at Korea Atomic Energy Research Institute (KAERI), and instructor at Seoul National University (Department of Nuclear Engineering) in Korea. Dr. Yim has been a consultant to the Electric Power Research Institute, Maine Yankee Atomic Power Co., Consumer Energy, the State of Washington, Korea Hydro and Nuclear Power Company, Korea Advanced Institute of Science and Technology, Energy Research Group, and Landauer, Inc. He is a member of the American Nuclear Society, the American Society for Testing and Materials, the Health Physics Society, Sigma Xi, Korean Nuclear Society, and Alpha Nu Sigma. He is also a member of ASME Peer Review Panel for DOE Office of Science and Technology and has been an invited reviewer for NRC Program Review Meetings. Dr. Yim received B.S. and M.S. in Nuclear Engineering from Seoul National University and Ph.D. in Nuclear Engineering from University of Cincinnati through Fulbright Scholarship. He also received S.M. in Environmental Health Science and Sc.D. in Radiological Health from Harvard University.